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Hou, Xiaolin

Publication date:
2011

Document Version
Early version, also known as pre-print

[Link back to DTU Orbit](#)

Citation (APA):
Hou, X. (2011). *AMS Analysis of 129I and its Application in Environmental and Oceanographic Tracer Studies*. Abstract from The 4th East Asia AMS Symposium, Tokyo, Japan.

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AMS Analysis of ^{129}I and its Application in Environmental and Oceanographic Tracer Studies

Xiaolin Hou^{1, 2*}

1 Risø National Laboratory for Sustainable Energy, Technical University of Denmark, DK-4000 Roskilde, Denmark

* Correspondence to: Xiaolin Hou; E-mail: xiho@risoe.dtu.dk;

Keywords: AMS; Iodine-129; Chemical speciation; Atmospheric process; Oceanographic tracer

1. Introduction

Anthropogenic ^{129}I discharged from the spent nuclear fuel reprocessing plants at Cap La Hague (France) and Sellafield (UK) has widely dispersed in the Nordic waters including the Arctic, as well as to the terrestrial environment through atmospheric dispersion. Due to the high solubility and long residence time of iodine in seawater, anthropogenic ^{129}I has become an ideal oceanographic tracer for investigating transport pathways and the exchange of water masses. Iodine is a redox sensitive element, existing mainly as iodide and iodate, with minor amounts of organic iodine in marine water. The chemical speciation analysis of anthropogenic ^{129}I in seawater can be used not only to investigate the marine geochemical cycle of iodine, but to detect any variation in the chemistry of a particular marine system, such as primary production. Due to the high volatility of iodine, and forming various active gaseous compounds in the atmosphere, iodine is an important element in the atmospheric process including primary particulate formation and ozone depletion. Anthropogenic ^{129}I can be employed as tracer for investigation these process.

2. Determination of environmental ^{129}I and its chemical speciation analysis

Although many methods can be used for measurement of ^{129}I , but only neutron activation analysis(NAA) and accelerator mass spectrometry (AMS) are sensitive enough for measurement of ^{129}I in environmental level. Of them, only AMS can be used to measure ^{129}I in samples without anthropogenic contamination in a $^{129}\text{I}/^{127}\text{I}$ ratio lower than 10^{-10} . AMS analysis of ^{129}I has been rapidly increased in the past few years with the increased numbers of AMS installed in many laboratories, and less isobaric interference in AMS and consequently easy to be measured even in a small AMS of only 0.5MV. To improve the measurement method of low level ^{129}I in less anthropogenic contaminated samples with low concentration of stable iodine.

In the past three years, a series carrier free method has been developed in Xi'an AMS center, which is consists of efficient separation of iodine from the samples matrices using combustion and ion exchange chromatography combined with a preparation of carrier free iodine target using co-precipitation of iodine as

AgI-AgCl. AMS measurement method was also optimized to be able to measure ^{129}I and ^{127}I signal in target with microgram level iodine.

Speciation analysis of ^{129}I can provide many information and useful for environmental tracer studies. In environmental water samples, iodine mainly exists as iodide, iodate and organic iodine, and in open seawater samples organic iodine is negligible, while in fresh water especially lake and river water, organic iodine might be a dominate species. A series speciation analysis method for ^{129}I in water samples have been developed in our lab. For seawater samples, anion exchange method is used to separated iodide and iodate, based on the high affinity of iodide to anion exchange resin comparing with iodate and other anions. Iodide on the column is eluted with high concentration of NaNO_3 solution, afterwards precipitated as AgI-AgCl co-precipitate for AMS measurement. Iodate in the effluent is reduced to iodide by NaHSO_3 in acidic solution, which is then separated using the same method as iodide for AMS measurement. For fresh water, a decomposition of organic iodine can be carried out using NaOH and NaClO solution, iodine in the treated water is then reduced to iodide using NaHSO_3 in acidic solution and separated using the same method as iodide. By this way, iodide, iodate and organic iodine can be determined. Recently, a new method has been developed to simplify the speciation analysis procedure of ^{129}I in seawater. Iodide and iodate is co-precipitated by AgI-AgCl using suitable amount of Ag^+ and NaHSO_3 in different pH values. The recoveries of iodide and iodate are higher than 80%, and cross-over contamination between iodide and iodate is less than 2%. This is very useful for rapis separation on board. The detailed analytical methods will be presented.

3. Application of ^{129}I as oceanographic tracer

Depth profiles of seawater were collected from more than 300 locations in the North Sea, Baltic Sea, Norwegian Sea, Arctic, and Atlantic as well as the Antarctic in many expeditions during 2005-2010. The speciation analysis for ^{129}I and ^{127}I in iodide, iodate, and total iodine forms have been carried out in most of samples. A volume of 0.2-2.0 liters of samples were used for the chemical separation of iodine species depending on the ^{129}I level. The separated ^{129}I is prepared as AgI or AgI-AgCl and measured using AMS, and ^{127}I of different species separated from each water sample was measured using inductively coupled plasma mass spectrometry (ICP-MS). The summary of the analytical results on the concentrations of chemical species of ^{129}I and ^{127}I as well as $^{129}\text{I}/^{127}\text{I}$ ratios of different species of iodine for these water samples will be presented. The spatial variations of ^{129}I , ^{127}I and their speciation in seawater in the investigated ears will be derived. Sources and transport pathways of ^{129}I in the Nordic seas and Baltic Sea, the transformation process of chemical species of iodine and their mechanism will be discussed. As an example, Fig.1 shows the distribution of ^{129}I in the North Sea.

4. Application of ^{129}I as atmospheric tracer

In the atmosphere, iodine exists in many forms and varies with time and location. Precipitation enriched iodine from the atmosphere during its formation in the atmosphere, therefore reflect the level of ^{129}I in the

atmosphere in an integral time scale during its transport and formation. We have collected atmospheric iodine in the form of particulate associated, inorganic gaseous iodine and organic gaseous iodine 1 meter above the surface in different locations with a distance of 0.1 km to 1000 km from the North Sea 1 in 2007. The collected iodine in different species was separated using combustion and prepared as AgI for AMS measurement of ^{129}I , ^{127}I is directly measured in the trap solution after combustion. The results show a clearly decreased ^{129}I level with the increased distance from the North Sea in a distance of less than 500 km. The direction of wind is an critical parameter determined the chemical species of ^{129}I and ^{127}I . The detailed results will be presented. Fig. 2 shows the variation of different species of ^{129}I with the distance to the North Sea.

A time series of precipitation samples has been collected in Denmark since 2000, the chemical speciation analysis s of ^{129}I and ^{127}I in these waters were carried out. It is surprised that ^{129}I and ^{127}I are in different chemical species in the precipitate. Iodide is the dominate species of ^{129}I , while ^{127}I is mainly in iodate form. The source term of ^{129}I and ^{127}I in the precipitation, as well as its formation process will be discussed.

Acknowledgement: Prof. Ala Aldahan and prof. Göran Possnert from Uppsala University were heavily involved in this work, from sampling, measurement of ^{129}I to the analysis of data. Dr. Justin Gwynn from Norwegian Radiation Protection Authority participated sampling and provided the seawater samples from Norwegian Sea and the Arctic, he has also involved in the analysis of data from these seawater samples. Prof. Weijian Zhou, Ning Chen, and Zhang Luyuan from Xi'an AMS centre, Institute of Earth Environmental were involved in the method development of AMS analysis of carrier free ^{129}I ; Maoyi Luo from Xi'an Jaotong University and Yukun Fan from Institute of Earth Environment, CAS participated in the analysis of seawater samples from the Arctic and Norwegian Sea. The work is partly supported by “BaiRen Project” (KZCX2-YW-BR-13) from CAS, China.

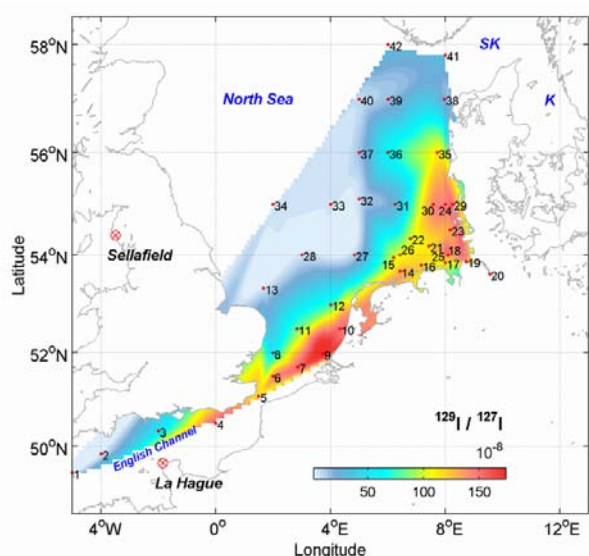


Fig. 1 Distribution of ^{129}I in the North Sea surface water collected in 2005

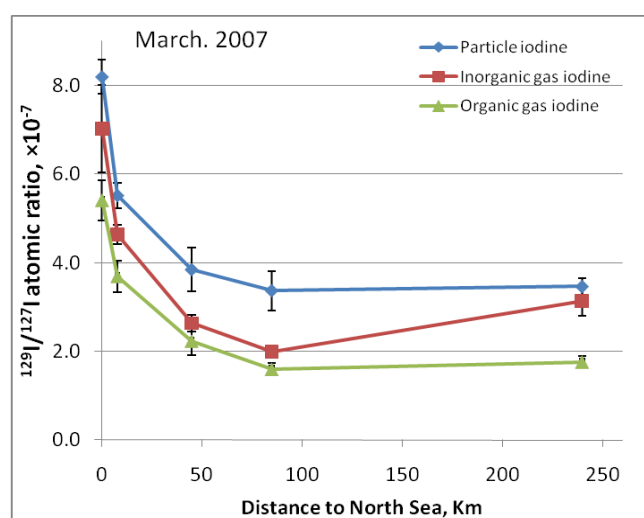


Fig. 2 Variation of ^{129}I species in the atmosphere collected in locations with different distance to the North Sea